Long-lived quantum memory with nuclear atomic spins

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We propose to store non-classical states of light into the macroscopic collective nuclear spin (10^{18} atoms) of a ^{3}He vapor, using metastability exchange collisions. These collisions, commonly used to transfer orientation from the metastable state 2^3S_1 to the ground state of ^{3}He, can also transfer quantum correlations. This gives a possible experimental scheme to map a squeezed vacuum field state onto a nuclear spin state with very long storage times (hours).

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If great progress has been made in the generation of non-classical states of light, a major challenge of quantum information and communication lies in the ability to manipulate and reversibly store such quantum states [1, 2]. Several proposals have been made to achieve storage of non-classical light states either in trapped cold atoms or atomic vapors [3, 4]. The first successful experiments of quantum memories for coherent states and squeezed states were achieved using atoms as a storage medium [5, 6]. In all the proposed schemes, as well as in the experiments realized so far, the information is encoded in the ground state of alkali atoms; the obtained storage times are at most several milliseconds, limited by collisions, magnetic field inhomogeneities, transit time, etc. Nuclear spins have also been proposed as quantum memories for mesoscopic systems, due to their long relaxation time [7]. In this Letter we show how to reversibly map a non classical state of light into a squeezed state, encoded in the purely nuclear spin of the ground state of ^{3}He, which interacts very little with the environment. The quantum state can then survive for times as long as *hours*. To access the ground state of ^{3}He, which is 20 eV apart from the nearest excited state, we propose to use metastability exchange (ME) collisions, during which an atom in the ground state and an atom in the metastable triplet state 2^3S_1 exchange their electronic variables. ME collisions are used in optical pumping of ^{3}He to create nuclear polarization in gas samples for nuclear physics experiments as well as in nuclear magnetic resonance imaging applications [8]. When the helium vapor is in a sealed cell, a weak radio-frequency discharge excited by a pair of external electrodes maintains a tiny fraction of the atoms in the metastable state, which has a finite lifetime due to its interactions with the cell walls. A transition is accessible from the metastable state to couple the metastable atoms with light. This, together with ME collisions, provides an effective coupling between the ground state atoms and light. We show that, with such a mechanism, quantum fluctuations can be reversibly transferred from the field to the atoms. Interacting with squeezed light in appropriate conditions, the macroscopic nuclear spin (1.6×10^{18} atoms of ^{3}He at 1 torr in a 50 cm^3 cell, at 300 K) of the polarized ground state gas becomes squeezed. The nuclear coherence relaxation time in absence of discharge and in an homogeneous field can be several hours. By switching on the discharge and repopulating the metastable state, the squeezing can be transferred back to the electromagnetic field and measured. In addition to its interest for quantum information, the scheme offers the exciting possibility to create a long-lived non classical state for spins.

We consider a system composed by N atoms in the ground state, and n atoms in the metastable state. These atoms interact with a coherent driving field with Rabi frequency Ω and frequency ω_1 that we treat classically, and a cavity field described by creation and annihilation operators A and $A^\dagger$ (Fig. 1-a). The field injected into the ring cavity, A_{in} with frequency ω_2, is in an amplitude-squeezed vacuum state: $\langle A_{in}\rangle = 0$ and $\Delta X_{in}^2 = e^{-2r}$, $\Delta Y_{in}^2 = e^{2r}$, where $X = A + A^\dagger$ and $Y = i(A^\dagger - A)$ are the standard field amplitude and phase quadrature operators, satisfying $[X, Y] = 2i$. The Hamiltonian of the atom-field system is:

$$H = H_0 + \hbar\left\{\Omega\, S_{31} e^{-i\omega_1 t} + g\, A\, S_{32} + \text{h.c.}\right\} \qquad (1)$$

where H_0 describes the free evolution of the atoms and the field, $g = d(2\pi\omega_2/\hbar V)^{1/2}$ is the coupling constant between the atoms and the cavity field, V being the volume of the cavity mode, d the atomic dipole. $S_{kl} = \sum_{i=1}^{n} |k\rangle_i\langle l|_i$ for $k, l = 1, 2, 3$ are collective atomic operators in the metastable and excited state [9]. The coupling to the ground state collective spin $I_{kl} = \sum_{i=1}^{N} |k\rangle_i\langle l|_i$ for $k, l = 9, 0$ is provided by ME collisions.

We start with a simplified picture, in which both the metastable and ground state atoms have a spin 1/2, which are simply exchanged during each ME collision. The exchange collisions rate for a metastable and a ground state atom are denoted by γ_m and γ_f, respectively. Their ratio γ_m/γ_f is equal to the ratio N/n. We assume that the system is initially prepared using optical pumping in the fully polarized state $\langle I_{00}\rangle = N$ and $\langle S_{22}\rangle = n$. Both the metastable and ground state collective spins are polarized along the z-axis of the Bloch sphere. The transverse spin components $S_x = (S_{21} + S_{21}^\dagger)/2$, $S_y = i(S_{21}^\dagger - S_{21})/2$ then play a similar role to field quadratures and, for such a coherent spin

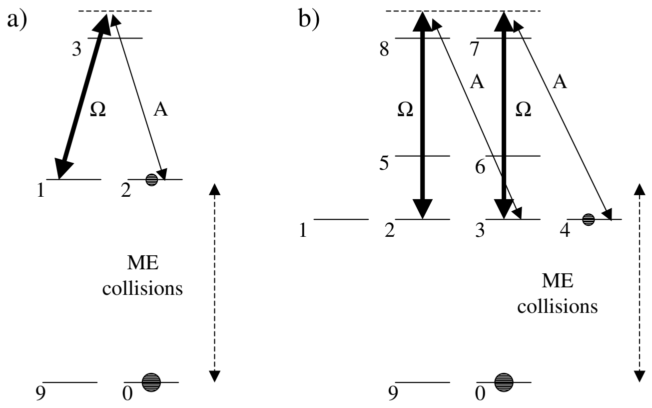

FIG. 1: a) Sublevels 1 and 2 are metastable, level 3 is the excited state, 9 and 0 are the ground state sublevels. b) Relevant energy levels in ^{3}He.

state, have equal variances: $\Delta S_x^2 = \Delta S_y^2 = n/4$ and $\Delta I_x^2 = \Delta I_y^2 = N/4$. By definition [10] the metastable (ground state) spin is squeezed if one of the transverse spin variance ΔS_x^2 or ΔS_y^2 (ΔI_x^2 or ΔI_y^2) is smaller than its coherent spin state value. As usual in quantum optics, we study the quantum fluctuations of operators around a "classical steady state" of the system (the fully polarized state). We then linearize the equations, and obtain in the rotating frame the following closed set of equations:

$$\dot{S}_{21} = -(\gamma_m - i\delta)S_{21} + \gamma_f I_{09} - i\Omega S_{23} + f_{21} \quad (2)$$
$$\dot{S}_{23} = -(\gamma + i\Delta)S_{23} - i\Omega S_{21} - ignA + f_{23} \quad (3)$$
$$\dot{I}_{09} = -(\gamma_f - i\delta_I)I_{09} + \gamma_m S_{21} + f_{09} \quad (4)$$
$$\dot{A} = -(\kappa + i\Delta_c)A - igS_{23} + \sqrt{2\kappa}A_{in} \quad (5)$$

We have introduced the detunings $\delta = \omega_S - \delta_{las}$, $\delta_I = \omega_I - \delta_{las}$, $\Delta = (E_3 - E_2)/\hbar - \omega_2$ with E_i the energy of level i, $\omega_I = (E_0 - E_9)/\hbar$, $\omega_S = (E_2 - E_1)/\hbar$, $\delta_{las} = \omega_1 - \omega_2$, and the cavity detuning $\Delta_c = \omega_c - \omega_2$. Ω is assumed to be real. The stochastic part of the evolution (quantum noise) of each operator is described by a time-dependent Langevin operator. If α and β denote two system operators, $\langle f_\alpha(t) f_\beta(t')\rangle = D_{\alpha\beta}\delta(t-t')$ where $D_{\alpha,\beta}$ is the corresponding coefficient of the diffusion matrix. Contributions to D come from polarization decay with a rate γ, ME collisions for metastable and ground state atoms, and cavity losses with a rate κ for the cavity field. The nonzero coefficients of the atomic part of the diffusion matrix are $D_{21,12} = D_{09,90} = 2n\gamma_m$, $D_{21,90} = D_{09,12} = -2n\gamma_m$, $D_{23,32} = 2n\gamma$, calculated using the generalized Einstein relation [11] for an ensemble of uncorrelated atoms. The Langevin forces for ME collisions are necessary for the model to be consistent. Otherwise the non Hamiltonian character of the exchange terms leads to violation of the Heisenberg uncertainty relations. Physically these forces originate from the fluctuating character of the ME collisions. Their correlation time is the collision time, much shorter than all the times scales we are interested in.

By adiabatic elimination of the polarization S_{23} and the cavity field assuming $\gamma, \kappa \gg \gamma_m, \gamma_f$, one obtains

$$\begin{aligned}\dot{S}_{21} + (\gamma_m + \Gamma - i\tilde{\delta})S_{21} &= \gamma_f I_{09} + f_{21} \\ &\quad - \frac{\Omega}{\Delta} f_{23} + i\frac{\Omega g n}{\Delta}\sqrt{\frac{2}{\kappa}} A_{in} \end{aligned} \quad (6)$$

where we introduced the optical pumping parameter $\Gamma = \gamma\Omega^2(1+C)/\Delta^2$, and the cooperativity $C = g^2 n/(\kappa\gamma)$, and we redefined the two-photon detuning $\tilde{\delta} = \delta + \Omega^2/\Delta$ to account for the light-shift of level 1. In deriving (6) we assumed a Raman configuration $\Delta \gg \gamma$, $\frac{C\gamma}{\Delta} \ll 1$ and that the cavity detuning exactly compensates the cavity field dephasing due to the atoms: $\Delta_C = C\kappa\gamma/\Delta$. Optimal coupling between the squeezed field and the metastable coherence is achieved under resonant conditions $\tilde{\delta} = 0$, or

$$\omega_S(B) + \Omega^2/\Delta = \omega_1 - \omega_2 \quad (7)$$

where the Larmor frequency ω_S can be adjusted using a magnetic field. A second resonance condition is $\delta_I = 0$, or

$$\omega_I(B) = \omega_1 - \omega_2 \quad (8)$$

meaning that the natural evolution frequency of the ground state coherence I_{09} should match that of the metastable state coherence. The Larmor frequency in the metastable and ground states are very different due to the difference between the nucleon and the electron mass. In low field, $\hbar\omega_\alpha = \mu_\alpha B$ (α=I,S) with $\mu_I/h = 3.24$kHz/G and $\mu_S/h = 1.87$MHz/G. However, the light shift in the metastable state allows to simultaneously fulfill (7) and (8) for a non zero magnetic field. Physically, these conditions ensure that both spin coherences are resonantly excited with the same tunable frequency $\omega_I(B)$, thus ensuring the efficiency of the squeezing transfer from the field to ground state. From Eq. (6) and the corresponding equation for I_{09} with $\tilde{\delta} = \delta_I = 0$, we can calculate the variances of the metastable and ground state spins. In the limit $\gamma_f \ll \Gamma, \gamma_m$ one obtains:

$$\Delta I_y^2 = \frac{N}{4}\left\{1 - \frac{\gamma_m}{\Gamma + \gamma_m}\frac{C}{C+1}(1 - e^{-2r})\right\} \quad (9)$$
$$\Delta S_y^2 = \frac{n}{4}\left\{1 - \frac{\Gamma}{\Gamma + \gamma_m}\frac{C}{C+1}(1 - e^{-2r})\right\}. \quad (10)$$

In the limit $C \gg 1$, the squeezing can be completely transferred to the atoms. If $\Gamma \gg \gamma_m$, correlations are established among the metastable-state spins, the leakage of correlation towards the ground state being negligible. The metastable collective spin is squeezed while the ground state spin remains unsqueezed. In the opposite limit $\Gamma \ll \gamma_m$, spin exchange is the dominant process for metastable atoms; they transfer their correlations to the ground state which then becomes squeezed. In all regimes the metastable and the fundamental state share the amount of noise reduction in the field.

In usual optical pumping experiments, the relevant atomic observables are the level orientations, i.e. one-body observables. ME collisions constantly tend to equal the degree of polarization of the two levels. By contrast, squeezing a spin component amounts to giving a negative value to the two-spin correlation function $\langle s_y(1)s_y(2)\rangle$. ME collisions constantly tend to equal the spin correlation functions in the two levels but not the degree of squeezing. This is because, to create maximum squeezing, a much smaller (absolute) value of the correlation function is needed in the fundamental than in the metastable state, due to the large population difference in the two levels [12]. Somehow paradoxically, the squeezed field then maintains a strong squeezing in the ground state via a weakly squeezed metastable state.

If one switches on the discharge and the coherent field in the same configuration as for the "writing" phase [4], the nuclear spin memory can be "read", the squeezing being transferred back to the electromagnetic field where it can be measured. During this process, the metastable level acquires only a weak degree of squeezing under the effect of ME collisions. But, because of the optical coupling, this squeezing progressively transits back to the quantum field stored in the cavity, so that, in the end, a strong squeezing is accumulated in the field without ever being large in the metastable state.

One important issue is the "writing" (or "reading") time of the quantum memory, which is the ground state effective response time. The adiabatic elimination of the metastable state in Eq. (4) shows that this time is the inverse of $\Gamma_F = \frac{\gamma_f\Gamma}{\gamma_m+\Gamma}$, which is also the width of the squeezing spectrum in the ground state.

We now apply our scheme to ^{3}He atoms in realistic conditions (Fig. 1-b). The coherent field (π-polarized) and the squeezed vacuum (σ^--polarized) are tuned to the blue side of the so called C_9 transition ($\lambda = 1.08\ \mu$m) from the $F = 3/2$ level of the 2^3S_1 metastable state to the $^3P_0$ state, the highest in energy of the 2^3P multiplicity [13]. The system is initially prepared in the fully polarized state, $\langle I_{00}\rangle = N$ and $\langle S_{44}\rangle = n$, by preliminary optical pumping. The metastable state now has two sublevels $F = 3/2$ and the $F = 1/2$. The effect of ME collisions on the metastable and ground state density matrices ρ_m and ρ_f can be written as [14]:

$$\begin{aligned}\dot{\rho_f} &= \gamma_f(-\rho_f + \mathrm{Tr}_e\rho_m)\\ \dot{\rho_m} &= \gamma_m(-\rho_m + \rho_f \otimes \mathrm{Tr}_n\rho_m)\end{aligned}$$

where Tr_e and Tr_n represent trace operations over the electronic and nuclear variables. After elimination of hyperfine coherences and linearization around the initially prepared state, we obtain a set of 11 closed equations involving the ground state coherence, the cavity field, 4 optical coherences, the excited state coherence, and 4 $\Delta m_F = 1$ coherences in the metastable state. To account for the fact that metastable atoms are destroyed as they reach the cell walls, we introduce a damping rate γ_0 of the metastable state coherences. Despite the more complicated level structure, in the fully polarized limit considered here, the squeezing transfer to the ground state comes exclusively from the coherence S_{34} which should be excited resonantly. By adiabatic elimination of the field and optical coherences, for optimal squeezing transfer conditions and in the limit $\gamma_f \ll \Gamma, \gamma_m$, we worked out the same analytical expressions (9-10) for the ground state and metastable spin variances, within a scaling factor in the optical pumping parameter

$$\Gamma = \gamma 3\Omega^2(1 + C)/\Delta^2\,, \qquad (11)$$

with now $\Delta = (E_7 - E_4)/\hbar - \omega_2$. In Fig. 2 we show the analytical predictions (9-10) and a full numerical calculation for realistic experimental parameters: a 1 torr vapor at 300 K, with $\gamma_m = 5\times10^6 s^{-1}$, and $\gamma = 2\times10^7 s^{-1}$, and a metastable atom density of 3.2×10^{10} atoms/cm^3 corresponding to a ratio $n/N = 10^{-6}$. The relaxation rate γ_0 is inversely proportional to the gas pressure (at 1 torr $\gamma_0 = 10^3 s^{-1}$). Deviations from the analytical formulas

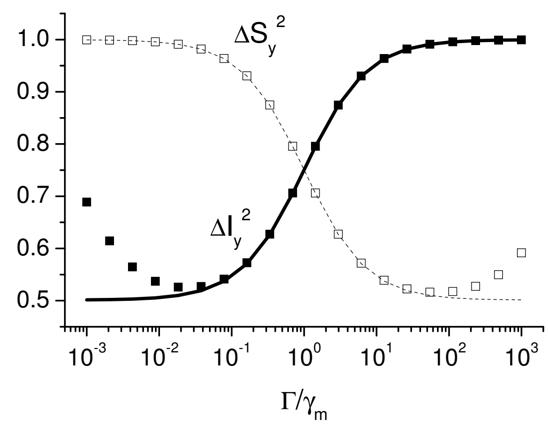

FIG. 2: Analytical predictions (lines) and numerical calculations for spin variances in ground state (full symbols) and metastable state (open symbols), as a function of the ratio Γ/γ_m. Numerical values of parameters are $e^{-2r} = 0.5$, $C = 500$, $\kappa = 100\gamma$, $\Delta = -2000\gamma$, $\gamma = 2\times10^7\mathrm{s}^{-1}$, $\gamma_m = 5\times10^6\mathrm{s}^{-1}$, $\gamma_0 = 10^3\mathrm{s}^{-1}$.

are due to non adiabaticity of the optical coherence with respect to metastable variables, which affects the squeezing of metastable spin, and to a finite relaxation rate in the metastable state γ_0, which affects the ground state spin squeezing in the region $\Gamma \ll \gamma_m$. In this figure the one-photon detuning Δ is kept fixed while the magnetic field and δ_{las} are chosen to satisfy simultaneously (7) and (8) with now $\omega_S = (E_4 - E_3)/\hbar$. The energy positions of atomic levels in the metastable and excited state depending on the field were computed including the effect of hyperfine interactions [15].

We calculated by simulation the effect of a frequency mismatch in (7) or (8), on spin squeezing in the ground

state. A frequency mismatch of the order of $\Gamma/3$ in the metastable state or of the order of Γ_F in the ground state affects the efficiency of the squeezing transfer. The condition for the ground state frequency matching (8) imposes stringent requirements on the homogeneity of the magnetic field [16]. Physically, if a significant dephasing between the squeezed field and the ground state coherence builds up during the squeezing transfer time, the atoms will see an average between the squeezed and the antisqueezed quadrature of the field, always above the standard quantum noise limit. Let ΔB be the maximum field difference with respect to the optimal value in the cell volume. For low field, the condition on ΔB to preserve the transfer efficiency reads $\mu_I \Delta B < h\Gamma_F$. Since $\Omega^2/\Delta \simeq \Gamma \frac{\Delta}{3\gamma C} \simeq \frac{\mu_S}{h} B$ we get $\frac{\Gamma}{\Gamma_F}\frac{\mu_I}{\mu_S}\frac{\Delta}{3\gamma C}\frac{\Delta B}{B} < 1$ or, in the regime $\Gamma \ll \gamma_m$, $600\frac{\Delta}{\gamma C}\frac{\Delta B}{B} < 1$. In Fig. 3 we show

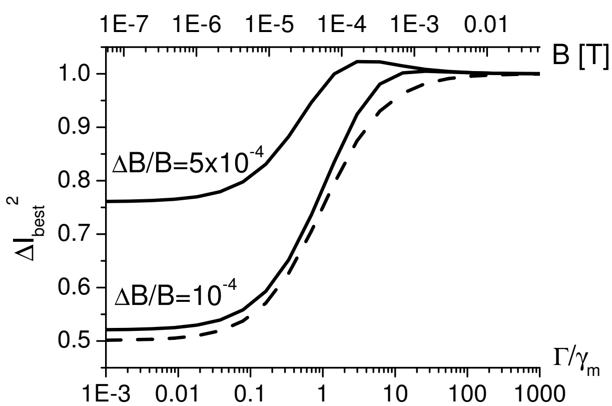

FIG. 3: Ground state spin quadrature optimized (best) variance as a function of the ratio Γ/γ_m (lower x-axis), for two relative changes of the magnetic field with respect to its optimal value (upper x-axis). $\Delta B/B = 0$ for the dashed curve. $\gamma_0 = 0$ and other parameters are as in figure 2.

the effect of a relative change of the magnetic field with respect to the optimal calculated value. An homogeneity of 100 ppm is sufficient for the chosen parameters to guarantee that all atoms are squeezed. The optimal calculated value for the field is shown as a second x-axis in the figure. In realistic conditions, choosing $\Gamma = 0.1\gamma_m$, the required field is about $B = 57$mG, corresponding to $\omega_I = 184$Hz. Squeezed vacuum states that can be generated for analysis frequencies as low as 200 Hz [17], could thus be efficiently transferred to the nuclear spins. The readout time is as long as the writing time: $\Gamma_F^{-1} = 2$s for $\Gamma = 0.1\gamma_m$, limited by the metastable atoms density in the sample.

The possibility to manipulate the spins using nuclear magnetic resonance techniques, and to optically readout the spin state after a long storage time makes this system particularly promising for quantum information [18].

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